

# PATENT SPECIFICATION

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## (54) UNSATURATED POLYESTERS AND THEIR USE AS BINDERS FOR GLASS FIBRES

(71) We, KAO SOAP CO. LTD., a Japanese Company, of 7-18, 1-chome, Nihonbashi-Bakurocho, Chuo-Ku, Tokyo, Japan, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed to be particularly described in and by the following statement:—

This invention relates to a binder for glass fibers, and to a method of producing glass fiber mats using such a binder.

Oil-in-water type emulsions of unsaturated polyester resins are used as binders for bonding glass fibers and preparing mat-like or pre-formed molded articles of glass fibers. Some of the binders are polymerized in the presence of an organic peroxide catalyst, whereas other binders are not polymerized in such a manner. The binder of this invention is of the latter type. According to this invention, glass fibers can be bonded, without employing either a vinyl monomer or a catalyst, by spraying an oil-in-water type emulsion of a polyester onto glass fibers and then removing water in an oven maintained at 100 to 200°C.

Binders for glass fibers are divided into the general classes of (1) the emulsion type and (2) the powder type, depending on the physical form in which they are applied. Use of an emulsion type binder is advantageous because the distribution of the binder on the glass fiber mat is more uniform and a more uniform fiber glass mat can be obtained. In the case of a powdery binder, in order to prevent undesired caking of the binder during storage, a resin having a high softening point should be used as a binder. As a consequence, the resulting glass fiber mat is relatively hard and this is disadvantageous because when the mat is folded, it is readily cut or broken at the fold line. However, when an emulsion type binder is employed, as is the case in this invention, this disadvantage does not occur. Because a resin having a low softening point can be used to impart flexibility to the resulting mat, even when the resulting mat is folded, the retention ratio of the tensile

strength is much higher than in the case of a glass fiber mat prepared using a powdery binder. In general, heretofore unsaturated polyesters have not been used as an emulsion type binder for the formation of glass fiber mats; rather homopolymers or copolymers of vinyl acetate have heretofore been used for this purpose. However, laminates of mats prepared using the prior art polyvinyl acetate-type emulsion binders are not transparent. Especially in the art of preparing glass fiber mats for corrugated plates, the development of binders giving a good transparency has been demanded, and polyvinyl acetate-type binders are insufficient in this point.

According to this invention, we provide a binder for glass fibers, comprising an unsaturated polyester in the form of an oil-in-water emulsion, which polyester has been obtained by the reaction of (a) 1 mole of an unsaturated dibasic acid component containing at least 50 mole percent of a material selected from fumaric acid, maleic anhydride and mixtures thereof, and (b) 1 to 1.5 moles of a diol component containing at least 30 mole percent of an adduct of 2 to 10 moles of an alkylene oxide to bisphenol A with (c) an emulsifier selected from an adduct of 5 to 200 moles of an alkylene oxide to castor oil, an adduct of 5 to 200 moles of an alkylene oxide to hydrogenated castor oil and polyethyleneglycol having an average molecular weight of 1000 to 6000, the amount of said emulsifier (c) being 5 to 30 parts by weight per 100 parts by weight of the sum of the polyester starting materials (a) and (b), the emulsion containing from 1% to 35% by weight of the polyester.

Glass fiber mats are generally prepared by cutting glass strands or filaments into prescribed lengths, piling the cut strands in random orientation to form a non-oriented fiber mat of a certain thickness and bonding the fibers with a binder.

The self-emulsifiable unsaturated polyester of the bisphenol type of this invention is formed by employing as a critical diol com-

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ponent (b) an alkylene oxide adduct to bisphenol A (the number of added moles of alkylene oxide being from 2 to 10) and reacting this adduct with an unsaturated dicarboxylic acid (a), in the presence of the emulsifier (c). It is possible to obtain unsaturated polyesters possessing different properties, namely, polyesters ranging from highly viscous liquids to solids having a softening point of 95°C., when the mixing ratio of (b) the alkylene oxide adduct to bisphenol A to the dibasic acid (a) is varied within the range of from 1 to 1.5 moles of (b) per 1 mole of (a).

In this invention, the thus-formed polyester is used in the state of an oil-in-water emulsion which is prepared by incorporating the polyester into water. The oil-in-water type polyester emulsion is especially useful as a binder for preparing glass fiber mats which are applied to fields in which molding is not conducted under high pressure. The concentration of the unsaturated polyester in the emulsion is in the range of 1 to 35 percent by weight, based on the total weight of the emulsion.

Glass fiber mats prepared with the binder of this invention possess unexpectedly improved properties in comparison with conventional products in that they have a sufficient tensile strength and even if they are once folded, they retain a very high proportion of their original tensile strength, and that laminates prepared from these glass fiber mats have very good transparency.

The oil-in-water type emulsion of an unsaturated polyester employed in this invention is prepared in the following manner.

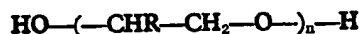
First, according to a conventional method, the reactants (a) and (b) are heated and reacted, in the presence of emulsifier (c) while removing the water formed by the polycondensation reaction. The reaction is carried out at a temperature of 150 to 210°C., preferably at 170 to 200°C. The reaction is preferably stopped when the acid value of the reaction mixture becomes lower than 30. Then, the resulting polyester is cooled to 100 to 150°C. and is gradually added, with sufficient agitation, to water maintained at 60 to 95°C., whereby the polyester is completely emulsified and an oil-in-water type emulsion of an unsaturated polyester is obtained.

It is critical that the unsaturated dibasic acid component (a), used for making the unsaturated polyester, consists of 50 to 100 mole percent of fumaric acid, maleic anhydride or mixtures thereof. The balance of the unsaturated dibasic acid component, that is, up to 50 mole percent thereof, can be selected from other  $\alpha,\beta$ -ethylenically unsaturated dibasic acids such as maleic acid and itaconic acid. It is industrially advantageous to employ an unsaturated dibasic acid component consisting essentially of 100 mole per-

cent of fumaric acid, maleic anhydride or mixtures thereof.

It is permissible to employ, as an optional additive, particularly if a softer plastics material is required, a small amount of up to 1 mole, per mole of said unsaturated dibasic acid component (a), of a saturated dibasic acid such as tetrahydrophthalic anhydride, phthalic anhydride, isophthalic acid, adipic acid and succinic acid, in combination with said unsaturated dibasic acid component (a).

It is critical to use as the diol component (b) of the polyester reaction mixture, a diol component consisting of 30 to 100 mole percent of an alkylene oxide adduct to bisphenol A. This adduct is formed by adding 2 to 10 moles of an alkylene oxide to 1 mole of bisphenol A (2,2 - bis(4 - hydroxyphenyl)propane)). As the alkylene oxide, there can be used, for example, ethylene oxide and propylene oxide. The balance of the diol component (b), if any, that is, up to 70 mole percent thereof, is (1) hydrogenated bisphenol A, (2) a diol of the following formula:



wherein n is an integer of from 1 to 4 and R is H or  $\text{CH}_3$ , or (3) mixtures of (1) and (2).

A diol of the above formula, hydrogenated bisphenol A and mixtures thereof can be used in combination with the essential constituent, the alkylene oxide adduct to bisphenol A. Specific examples of diols of the above formula include ethyleneglycol, propyleneglycol, diethyleneglycol, triethyleneglycol, tetraethyleneglycol, dipropyleneglycol, tripropyleneglycol and tetrapropyleneglycol.

As the emulsifier, there is used an alkylene oxide adduct to castor oil or hydrogenated castor oil, formed by adding 5 to 200 moles, preferably 50 to 150 moles, of an alkylene oxide, such as ethylene oxide or propylene oxide, to castor oil or hydrogenated castor oil. Ethylene oxide is generally employed as the alkylene oxide. Another emulsifier that is used is polyethyleneglycol having an average molecular weight of 1000 to 6000. Mixtures of these emulsifiers can be used.

The unsaturated polyester, according to this invention, is prepared by reacting 1 to 1.5 moles of the diol component (b), as defined above, with 1 mole of the dibasic acid component (a), as defined above. This reaction is carried out in the presence of the emulsifier (c).

The amount of the emulsifier, as defined above, used for preparing an unsaturated polyester is from 5 to 30 parts by weight of emulsifier per 100 parts by weight of the sum of the above defined dibasic acid component [the above component (a)] plus the

diol component [the above component (b)]. It is possible to add minor amounts of polymerization initiator, and defoaming agent to the reaction mixture according to need, in a conventional manner.

The oil-in-water emulsion containing the unsaturated polyester is applied to a layer of glass fibers in any conventional way, such as by spraying. This operation can be repeated as often as needed to form a mat of the desired thickness and then the mat is dried, such as in an oven heated to from 100°C. to 200°C., or in air, to remove the water whereby the glass fibers are adhered together by the unsaturated polyester. The amount of unsaturated polyester applied as binder is generally in the range of 3 to 7 weight percent (calculated on a water-free basis), based on the weight of the glass fibers.

According to this invention, glass fiber mats having a good transparency and a high tensile strength retention ratio after folding can be obtained by using the oil-in-water type polyester emulsion prepared under the above-described specific conditions. These glass fiber mats have a very high practical utility.

#### Example 1

A reaction vessel equipped with an agitator, a thermometer, a nitrogen gas inlet tube and a water outlet tube was charged with 1050 g. (3.0 moles) of 2.0 prodendro-bisphenol A (adduct of 2.0 moles of propylene oxide to bisphenol A), 323 g. (2.8 moles) of fumaric acid, 320 g. of polyethyleneglycol #1500 (having an average molecular weight of about 1500), 1.0 g. of hydroquinone as a polymerization inhibitor and 0.3 g. of silicone (marketed under the tradename "Toshiba Silicone TSA-730") as a defoaming agent, and the mixture was heated and reacted at 160 to 220°C., in a nitrogen gas current. The reaction was stopped when the acid value of the formed polyester was 25. The reaction product was cooled to 100°C. and then was gradually poured with agitation into water maintained at 70 to 80°C. There was obtained a homogeneous unsaturated polyester emulsion. The water content of the thus-obtained oil-in-water type unsaturated polyester emulsion was adjusted so that the polyester concentration thereof was 5 percent by weight.

Using the thus-obtained oil-in-water type unsaturated polyester emulsion, fiber glass mats were prepared according to the following method.

A perforated steel plate having a size of 35 cm×35 cm was mounted on a mat-preparing test machine provided with a blower, and 20 g. of chopped glass fibers was uniformly and randomly scattered from above to form a first glass fiber layer on the steel plate. Then, 30 g. of the oil-in-water type unsaturated polyester emulsion having a polyester con-

centration of 5 percent by weight was sprayed on the layer. This procedure was repeated three times to form a glass fiber mat having a three-layer structure. Then, the mat was placed for 15 minutes in an oven maintained at 180°C. to remove water therefrom.

The resulting glass fiber mat was inserted between two steel plates and was cooled to room temperature in that state. Then it was cut into a size suitable for the styrene solubility test and tests were performed for determining the tensile strength and the fold tensile strength (the tensile strength after folding) thereof.

The styrene solubility of the glass fiber mat was determined in the following manner.

The glass fiber mat was cut into a size of 12.5 cm. length×10 cm. width, and the upper and lower edges thereof were pinched and secured by clips having substantially the same sizes with that of said upper and lower edges. A weight was hung on the lower clip so that the load of the clip and the weight was 100 g.±1 g. The upper clip was manually picked up and the test piece was immersed in a tank filled with styrene maintained at 25°C. Then, the upper clip was fixed to a supporting rod. Thus, the test piece was allowed to stand in styrene maintained at 25°C. under a load of about 100 g., and the time required for the sample glass fiber mat to break apart due to dissolution of the binder into styrene was measured.

The tensile strength was determined on a test specimen cut into a size of 10 cm.×25 cm. by means of a tensile tester (autographic tensile tester manufactured by Shimadzu Seisakusho).

The fold tensile strength was determined in the following manner.

The glass mat was cut into a size of 10 cm.×25 cm., and it was folded at an angle of 180° along the center line spaced 12.5 cm. from both the shorter sides of the cut sample. Then, the folded sample mat was spread flat to the original state, and the tensile strength was measured in the same manner as described above.

The tensile strength retention ratio after folding was calculated from the following formula:

$$\text{Tensile strength retention ratio (\%)} = \left[ \frac{\text{tensile strength after folding}}{\text{tensile strength before folding}} \right] \times 100$$

Each of the foregoing tests was conducted on 8 samples, and an average value was adopted for evaluation.

The results of these measurement tests made on the glass fiber mat prepared by employing the above-mentioned oil-in-water type polyester emulsion are as follows:

styrene solubility test: 4 minutes and 30 seconds  
 tensile strength: 21.1 Kg.  
 fold tensile strength: 17.9 Kg.  
 tensile strength retention ratio after folding: 84.8%

Plates prepared by impregnating the thus-obtained mat with commercially available unsaturated polyester resins, namely Atlac 382-05 (Registered Trade Mark—bisphenol A type manufactured by Kao Atlas), Polymar TG 3302 (phthalic acid type manufactured by Takeda Yakuhin Kogyo) and Polymar TH 302 (isophthalic type manufactured by Takeda Yakuhin Kogyo), and molding the impregnated mat had a very excellent transparency.

#### Example 2

The same reaction vessel as used in Example 1 was charged with 1037 g. (3.2 moles) of an adduct of 2 moles of ethylene oxide to bisphenol A, 294 g. (3.0 moles) of maleic anhydride, 330 g. of an adduct of 100 moles of ethylene oxide to castor oil, 1 g. of hydroquinone and 0.3 g. of silicone (marketed under the tradename "Toshiba Silicone TSA-730") as a defoaming agent, and the mixture was heated and reacted at 160 to 200°C. in a nitrogen gas current. A product having an acid value of 25 was obtained by conducting the reaction for about 6 hours. Then, the product was cooled to 100°C. and gradually poured under agitation into water maintained at 80 to 90°C. to obtain a homogeneous emulsion. The water content of the thus-obtained oil-in-water type unsaturated polyester emulsion was adjusted so that the polyester concentration was 5 percent by weight.

A glass fiber mat having a three-layer structure was prepared by using as a binder the thus-obtained oil-in-water type polyester emulsion according to the same method as described in Example 1.

The resulting mat was tested according to the methods described in Example 1, to obtain the following results:

styrene solubility: 5 minutes and 12 seconds  
 tensile strength: 18.1 Kg.  
 fold tensile strength: 14.9 Kg.  
 tensile strength retention ratio after folding: 82.3%

Plates prepared by impregnating the thus-obtained mat with the same commercially available unsaturated polyester resins as used in Example 1 and molding the impregnated mats, had a very excellent transparency.

#### Example 3

The same reaction vessel as used in Example 1 was charged with 567 g. (1.6 moles) of an adduct of 2 moles of propylene oxide to

bisphenol A, 580 g. (2.8 moles) of hydrogenated bisphenol A, 488 g. (4.2 moles) of fumaric acid, 333 g. of polyethyleneglycol having an average molecular weight of 4000, 1.4 g. of hydroquinone and 0.5 g. of silicone (marketed under the tradename "Toshiba Silicone TSA-730") as a defoaming agent, and mixture was heated and reacted at 160 to 210°C. in a nitrogen current for about 10 hours to obtain a product having an acid value of 29 and a softening point of 92°C. The thus-obtained reaction product was cooled to 140°C. and gradually poured under agitation into hot water maintained at 90 to 95°C. to obtain a homogeneous emulsion. The thus-obtained oil-in-water type unsaturated polyester emulsion was adjusted so that the polyester concentration was about 5 percent by weight.

A glass fiber mat having a three-layer structure was prepared by employing as a binder the thus-obtained oil-in-water type polyester emulsion according to the same method as described in Example 1.

The thus-obtained mat was tested according to the methods described in Example 1 to obtain the following results:

styrene solubility test: 4 minutes and 50 seconds  
 tensile strength: 16.8 Kg.  
 fold tensile strength: 12.1 Kg.  
 tensile strength retention ratio after folding: 72.0%

Plates prepared by impregnating the thus-obtained mat with the same commercially available unsaturated polyester resins as used in Example 1 and molding the impregnated mat, had a very excellent transparency.

#### Example 4

The same reaction vessel as used in Example 1 was charged with 1128 g. (3.2 moles) of an adduct of 2 moles of propylene oxide to bisphenol A, 76 g. (1.0 mole) of propyleneglycol, 314 g. (3.2 moles) of maleic anhydride, 120 g. (0.8 mole) of phthalic anhydride, 427 g. of an adduct of 100 moles of ethylene oxide to hydrogenated castor oil, 1.6 g. of hydroquinone and a very small amount of silicone (marketed under the tradename "Toshiba Silicone TSA-730"), and the mixture was heated and reacted at 160 to 210°C. in a nitrogen current for about 11 hours to obtain a product having an acid value of 24. Then, the thus-obtained reaction product was cooled to 100°C. and gradually poured under agitation into water maintained at 80 to 90°C. to obtain a homogeneous emulsion. The thus-obtained oil-in-water type unsaturated polyester emulsion was adjusted so that the polyester concentration was 5 percent by weight.

A glass fiber mat having a three-layer structure was prepared by using as a binder the

thus-obtained oil-in-water type polyester emulsion according to the method described in Example 1.

The thus-obtained mat was tested according to the methods described in Example 1 to obtain the following results:

styrene solubility test: 4 minutes and 10 seconds  
tensile strength: 18.5 Kg.  
fold tensile strength: 15.9 Kg.  
tensile strength retention ratio after folding: 85.9%

Plates prepared by impregnating the thus-obtained mat with the same commercially available unsaturated polyester resins as used in Example 1 and molding the impregnated mat, had a very excellent transparency.

#### Comparative Example 1

A reaction vessel was charged with 688 g. (6.5 moles) of diethyleneglycol, 697 g. (6.0 moles) of fumaric acid, 277 g. of polyethyleneglycol having an average molecular weight of 1540, 1.5 g. of hydroquinone and 0.6 g. of silicone (marketed under the tradename "Toshiba Silicone TSA-730") as a defoaming agent, and the mixture was heated and reacted at 160 to 210°C. in a nitrogen current for about 8 hours to obtain a product having an acid value of 25. The thus-obtained reaction product was cooled to 100°C. and gradually poured under agitation into water maintained at 70 to 80°C. to obtain an emulsion. The thus-obtained oil-in-water type unsaturated polyester emulsion was so adjusted that the polyester concentration was 5 percent by weight.

A glass fiber mat having a three-layer structure was prepared by employing as a binder the thus-obtained oil-in-water type polyester emulsion according to the same method as described in Example 1.

The thus-obtained mat was tested according to the methods described in Example 1 to obtain the following results:

styrene solubility test: 4 minutes and 55 seconds  
tensile strength: 6.8 Kg.  
fold tensile strength: 4.8 Kg.  
tensile strength retention ratio after folding: 70.6%

Plates were prepared by impregnating the thus-obtained mat with the same commercially available unsaturated polyester resins as used in Example 1 and molding the impregnated mat. Each of the thus-obtained plates was whitish and therefore opaque.

#### Comparative Example 2

A glass fiber mat having a three-layer structure was prepared by employing, instead of

the oil-in-water type unsaturated polyester emulsion of Example 1, a commercially available powdery binder resin (marketed under the tradename "Atlas 363 E"; product of Kao Atlas). More specifically, in the same manner as described in Example 1, 20 g. of chopped glass strand was uniformly and randomly scattered on the steel plate, and 1.5 g. of the above powdery binder was uniformly scattered on the chopped glass strand. Then 5 g. of water was uniformly scattered thereon. This procedure was repeated three times, and water was removed by heating, to obtain a mat having a three-layer structure.

The thus-obtained mat was tested according to the methods described in Example 1 to obtain the following results:

styrene solubility test: 3 minutes and 10 seconds  
tensile strength: 17.9 Kg.  
fold tensile strength: 7.6 Kg.  
tensile strength retention ratio after folding: 42.4%

Plates were prepared by impregnating the thus-obtained mat with the same commercially available unsaturated polyester resins as used in Example 1 and molding the impregnated mats. The resulting plates had a fair transparency but they were inferior in this respect to the plates obtained in Examples 1 to 4.

#### WHAT WE CLAIM IS:—

1. A binder for glass fibers, comprising an unsaturated polyester in the form of an oil-in-water emulsion, which polyester has been obtained by the reaction of (a) 1 mole of an unsaturated dibasic acid component containing at least 50 mole percent of a material selected from fumaric acid, maleic anhydride and mixtures thereof, and (b) 1 to 1.5 moles of a diol component containing at least 30 mole percent of an adduct of 2 to 10 moles of an alkylene oxide to bisphenol A with (c) an emulsifier selected from an adduct of 5 to 200 moles of an alkylene oxide to castor oil, an adduct of 5 to 200 moles of an alkylene oxide to hydrogenated castor oil and polyethyleneglycol having an average molecular weight of 1000 to 6000, the amount of said emulsifier (c) being 5 to 30 parts by weight per 100 parts by weight of the sum of the polyester starting materials (a) and (b), the emulsion containing from 1% to 35% by weight of the polyester.

2. A method of producing glass fiber mats in which a polyester resin in the form of an oil-in-water emulsion according to Claim 1 is applied as a binder to glass fibers to form a mat.

3. A binder according to Claim 1, in which a part of the dibasic acid component (a) is an  $\alpha,\beta$ -ethylenically unsaturated dibasic acid

other than fumaric acid and up to 70 mole percent of the diol component is selected from hydrogenated bisphenol A, a diol of the formula



wherein  $n$  is an integer from 1 to 4 and  $R$  is  $\text{H}$  or  $\text{CH}_3$ , and mixtures thereof, and which additionally contains up to 1 mole, per mole of component (a), of a material selected from tetrahydrophthalic anhydride, phthalic anhydride, isophthalic acid, adipic acid, succinic acid, and mixtures thereof, the acid value of the polyester being lower than 30.

4. A method of making a binder according to Claim 3 in which said binder is obtained by reacting at a temperature of 150 to 210°C., a reaction mixture consisting of components (a), (b), and (c) and (d) wherein the mole ration of

$$20 \quad \frac{(b)}{(a)}$$

or

$$\frac{(b)}{(a) \text{ plus } (d)}$$

is

$$\frac{1.0 \text{ to } 1.5}{1.0}$$

25 and the weight ration of

$$\frac{(c)}{(a) \text{ plus } (b)}$$

or

$$\frac{(c)}{(a) \text{ plus } (b) \text{ plus } (d)}$$

is

$$30 \quad \frac{5 \text{ to } 30}{100}$$

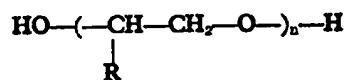
wherein component (a) consists of (1) 50 to

100 mole percent of a material selected from fumaric acid, maleic anhydride and mixtures thereof, and

(2) the balance of (a) is an  $\alpha,\beta$ -ethylenically unsaturated dibasic acid other than fumaric acid, component (b) consists of

(1) 30 to 100 mole percent of an adduct of 2 to 10 moles of alkylene oxide to bisphenol A, and

(2) the balance is selected from hydrogenated bisphenol A, a diol of the formula



wherein  $n$  is an integer from 1 to 4 and  $R$  is  $\text{H}$  or  $\text{CH}_3$ , and mixtures thereof

component (c) is selected from an adduct of 5 to 200 moles of an alkylene oxide to castor oil, an adduct of 5 to 200 moles of an alkylene oxide to hydrogenated castor oil and polyethylene glycol having an average molecular weight of 1000 to 6000, and

component (d) is up to 1 mole, per mole of said component (a), of a material selected from tetrahydrophthalic anhydride, phthalic anhydride, isophthalic acid, adipic acid, succinic acid and mixtures thereof

the reaction being carried out until the acid value of the reaction mixture is less than 30, and the reaction product then being made up into an emulsion with water.

5. A method according to Claim 4, wherein said alkylene oxide is ethylene oxide or propylene oxide.

6. A method according to Claim 5, wherein component (a) consists of fumaric acid.

7. A method according to Claim 5, wherein component (a) consists of maleic anhydride.

8. A method according to Claim 2, wherein said binder is sprayed onto said glass fibers to deposit thereon from 3 to 7 weight percent of said polyester, calculated on a water-free basis, based on the weight of the glass fibers, and then drying said mat in an oven at 100 to 200°C until said mat is substantially water-free.

9. A method of producing glass fiber mats substantially as herein described with reference to Examples 1 to 4.

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